Regenerated Uranium Separation in Matched Abundance Ratio Cascade with Additional Product Flow

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Regenerated Uranium Separation in Matched Abundance Ratio Cascade with Additional Product Flow

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Abstract. This paper addresses known methods used to purify regenerated uranium in single and double cascades. A new method for separating regenerated uranium has been developed that enables a significant reduction of the concentration of $^{232, 234}$U in the additional product flow. Matched abundance ratio cascades (M*-cascades) with different key components and additional product flow are used in the new method. Main product flow of the M*-cascade contains low enriched regenerated uranium. It can be used for reactor fuel production. Purified product can be enriched in the ordinary cascade in compliance with the requirements of ASTM C 996–10 set for isotopes $^{232, 234}$U in low enriched commercial uranium, which is usually produced from the natural one. Computer experiment based on the new method has been performed. The experiment shows that the best cascade with the maximum flow of the enriched purified product is M*-cascade with key components ^{232,236}U.

1. Introduction

Regenerated uranium is produced from irradiated nuclear reactors fuel. It plays an important role in the nuclear fuel cycle. It contains isotope ^{235}U, which has a concentration higher than that in the natural uranium. However, there are significant amounts of impurity isotopes $^{232, 234, 236}$U in the regenerated uranium. It is impossible to separate them chemically. The impurity isotopes cause the main difficulties for using regenerated uranium in the production of reactor fuel [1].

The most significant problem is the presence of the isotopes ^{232}U and ^{234}U because of their high radioactivity. The isotope ^{236}U captures neutrons in a nuclear reactor. Its presence leads to an increase of the concentration of the fissile isotope ^{235}U in the fuel. This problem affects the economic indicators of a nuclear reactor. To minimize radiation hazards and improve fuel quality it is necessary to reduce the concentration of $^{232, 234, 236}$U. For this purpose cascade separation technique of the uranium hexafluoride (UF_6) and dilution operations can be used.

There are different ways of regenerated uranium enrichment with the reduction of $^{232, 234, 236}$U concentration in a separate cascade. One of these is its enrichment to a concentration of the ^{235}U up to 10–90% in the ordinary cascade and subsequent dilution down to 2–7% [2]. Despite the good quality of the product obtained, this method has significant drawbacks: ^{235}U isotope with a concentration over 5% is formed as an intermediate product of the purification process. Moreover, this method involves using natural uranium.

Also, there are methods of regenerated uranium purification in double cascades [3–5]. The first ordinary cascade enriches the uranium hexafluoride up to 20% and above for ^{235}U. The product flow of this cascade is fed the second cascade, where $^{232, 234}$U are removed from the waste flow. The purified wasted uranium hexafluoride of the second cascade is diluted to obtain the desired concentration of ^{235}U. When the additional carrier gas [5] is used in the second cascade feed flow the content of ^{232}U is reduced down to 3×10^{-8} – 4×10^{-9} % without using special diluents. The only disadvantage of the double cascades [3–5] is obtaining a highly enriched uranium with ^{235}U concentration more than 20% at some stages of separation.

Another method uses additional feed flow with natural uranium in the cascade [6]. When using this method, the regenerated uranium is enriched to a concentration less than 5% ^{235}U while the content of $^{232, 234, 236}$U is decreased. The consumption of natural uranium can be efficiently reduced by utilizing one more feed flow with depleted waste uranium [7]. However, this leads to an increase in the expenses for separation work. Another method involves using a cascade with an additional product flow and two feed flows with regenerated and with natural uranium [8]. In this case, additional expenses for separation work are practically nonexistent. However, with a significant reduction in the concentration of $^{232, 234, 236}$U in the intermediate product flow, their content increases in the final commercial product flow of the low enriched uranium.

The alternative is a double cascade, where ^{235}U concentration is limited and the pattern is changed so as to remove $^{232, 234}$U in the first ordinary cascade. Thus, the feeding point is significantly shifted toward the product flow. Its purified depleted stream is fed to the second ordinary cascade, where the desired commercial product of low enriched uranium is generated in the product flow [9]. Studies have shown that reducing the concentration of ^{232}U to $(3\text{-}4)\times10^{-9}$ % in the final product is possible with ^{235}U enrichment to 5 % in the cascades [10]. However, this method features high expenses for separation work and does not provide for significant removal of $^{234, 236}$U isotopes. More efficient removal of ^{234}U is possible using the method described in [11].

This paper deals with the purification of the regenerated uranium in the additional product flow of the matched abundance ratio cascade (M*-cascade) [12] and subsequent enrichment of the purified product in the ordinary cascade. Calculations are based on the methodology [13] for the M*-cascades for various key components.

2. Calculations

The scheme consisting of two cascades is shown in Figure 1.

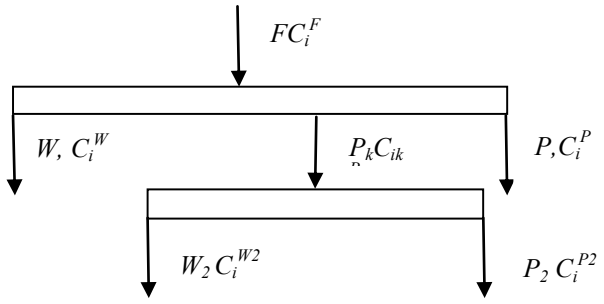

Figure 1. Double cascade scheme

Let F, W, and P denote feed, waste, and primary product flows of the first cascade, respectively; let P_k denote additional product flow, and C_i^F, C_i^W, C_i^P, $C_{ik}{}^P$ denote corresponding concentrations (mass fractions) of the i-th component. Additional flow P_k is taken from the waste flow of stage k of the first cascade. W_2, P_2 are waste and product flows of the second cascade; and C_i^{W2}, C_i^{P2} are the corresponding concentrations of the i-th component.

Matched abundance ratio cascades with one additional product flow were calculated for various key components. Feed flow F was 10 g/s and consisted of a mixture of isotopes: ^{232}U – 1.5×10^{-7} %, ^{234}U – 1.6×10^{-2} %, ^{235}U – 0.85 %, ^{236}U – 0.35 %.

The selected concentrations of feed correspond to the characteristic isotopic composition of regenerated uranium [14]. The cascades consisted of 51 stages with the feed in the stage number 45. This configuration has been chosen based on the given separation factors and the need to achieve a concentration of the ^{235}U not exceeding 5% in the primary product flow and 0.04 – 0.3% in the waste. Under these conditions, the primary product flow meets the requirements of ASTM C 996–10 for low enriched regenerated uranium and can be used to produce the corresponding reactor fuel.

The number of the additional product flow stage k varied from 2 to 45, its flow quantity P_k ranged from 1 g/s to 5 g/s in steps of 0.1 g/s. The separation factor per unit mass difference was assumed to be 1.091.

For each calculation, the additional product flow of the M*-cascade fed the ordinary cascade. The parameters of the latter were assessed by analytical formulas [15]. The concentration of the ^{235}U at this cascade was set equal to 0.3% for the waste and 4.4% for the product. The concentration of the ^{235}U in the product flow of the ordinary cascade was taken on the basis of its required content of 3.2 – 3.6% in the fuel without regard for the parasitic neutron capture by ^{236}U isotope. The compensation factor in the product was 0.2 – 0.3. The content of the impurity isotopes in the product was limited to 1×10^{-8} % for the ^{232}U and 4.8×10^{-2} % for the ^{234}U. These concentrations of impurity isotopes meet the ASTM C 996-10 specification for low enriched uranium with a concentration of commercial ^{235}U equal to 4.4%.

3. Results

It was found that the concentration of the components $^{232\text{-}236}$U decreases with the increase of the additional product flow (Table 1).

Table 1. Concentration of components in function of additional product flow for stage No. 14 (k = 14) of M*-cascade with key components $^{232,\,236}$U.

P_k(g/s)	C_{ik}^{P}(%)			
	^{232}U	^{234}U	^{235}U	^{236}U
1.7	6.45×10^{-10}	7.03×10^{-3}	0.8498	0.4057
2.0	6.32×10^{-10}	6.70×10^{-3}	0.8193	0.3978
2.5	6.12×10^{-10}	6.21×10^{-3}	0.7731	0.3853
3.0	5.94×10^{-10}	5.79×10^{-3}	0.7318	0.3736
3.5	5.76×10^{-10}	5.42×10^{-3}	0.6946	0.3626
4.0	5.60×10^{-10}	5.09×10^{-3}	0.6610	0.3521
4.4	5.47×10^{-10}	4.86×10^{-3}	0.6364	0.3442

As the additional product stage number increases, the $^{232\text{-}236}$U concentrations increase too (Table 2). It follows from these that an optimal stage number exists for each selected value of the additional product flow P_k in the matched abundance ratio cascade. The product flow of the ordinary cascade $P2$ will be maximal for this stage number taking into account the limits assumed.

Table 2. Concentration of components in function of additional product flow stage number of M*-cascade with key components ^{232,236}U when P_k=2.7 g/s.

k	C_{ik}^{P}(%)			
	^{232}U	^{234}U	^{235}U	^{236}U
2	5.58×10^{-11}	2.66×10^{-3}	0.469	0.287
5	1.07×10^{-10}	3.54×10^{-3}	0.568	0.324
10	2.87×10^{-10}	4.96×10^{-3}	0.691	0.364
14	6.05×10^{-10}	6.03×10^{-3}	0.756	0.381

It should be noted that when the restrictions on the concentration of ^{232,234}U are met the ^{236}U content in the product flow of the ordinary cascade increases as P_k grows (Figure 2).

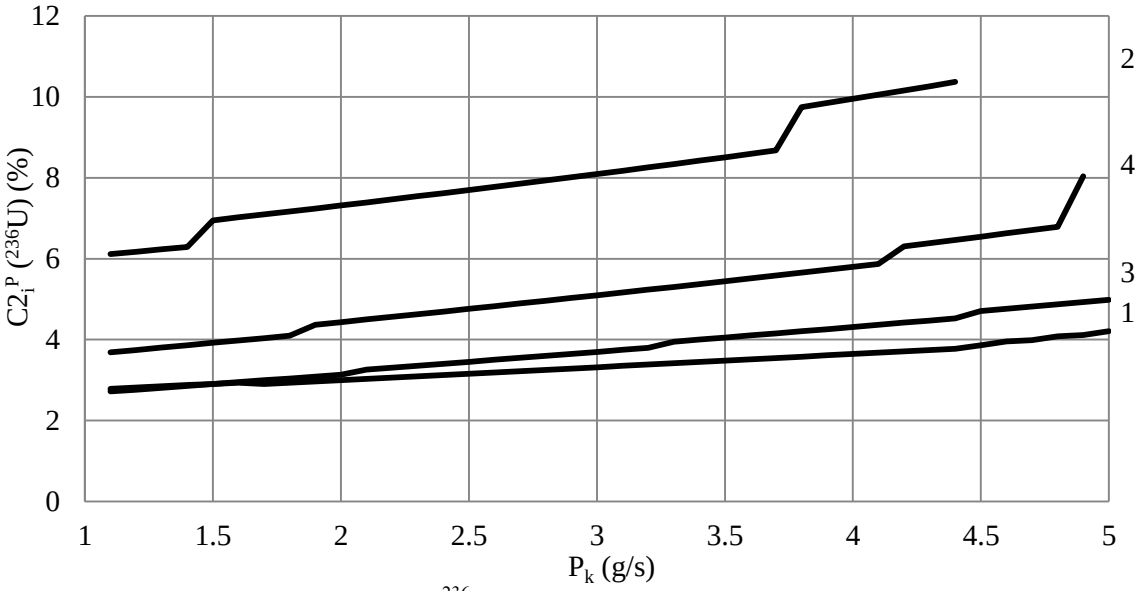

Figure 2. Concentration of the ^{236}U in product flow of ordinary cascade in function of additional product flow of M*-cascade: 1 – key components ^{232,236}U; 2 – key components ^{232,238}U; 3 – key components ^{234,235}U; 4 – key components ^{235,236}U.

Thus, it is possible to reduce the product flow of the ordinary cascade to decrease the ^{236}U concentration. For example, a decrease of the flow by 4 % from the optimal value for the matched abundance ratio cascade with key components ^{232,238}U leads to a decrease of the ^{236}U concentration by more than 16 % (Table 3).

Table 3. Variation of ^{236}U concentration in product flow of ordinary cascade

K	P_k(g/s)	C_{ik}^P(%)				$P2$ (g/s)	$C2_i^P$(%)		
		^{232}U	^{234}U	^{235}U	^{236}U		^{232}U	^{234}U	^{236}U
30	1.8	2.26×10^{-9}	4.40×10^{-3}	0.654	0.390	0.156	9.93×10^{-9}	3.76×10^{-2}	4.10
29	2.7	1.74×10^{-9}	3.50×10^{-3}	0.523	0.340	0.162	8.98×10^{-9}	3.96×10^{-2}	5.10

When the P_k flow increases the feed flow of the ordinary cascade also increases. At the same time, the ^{235}U concentration in the feed flow decreases. These two factors have opposite effects on generation of the product flow $P2$ at the given concentrations for the product and waste of the ordinary cascade. The first of them results in the increase of $P2$ and the second one leads to its decrease. As a result, the maximum $P2$ is observed at certain optimum value of P_k (Figure 3).

Table 4 presents examples of additional product flows of the M*-cascades that correspond to the maximal values of $P2$ with the restrictions set for ^{232,234}U met.

Table 4. Parameters of additional product flows of M*-cascades and ordinary cascades with maximal product flows $P2$.

MARC	k	P_k (g/s)	ΣL (g/s)	C_{ik}^P (%)				C_i^{P235} (%)	$P2$ (g/s)	$C2_i^P$ (%)		
				^{232}U	^{234}U	^{235}U	^{236}U			^{232}U	^{234}U	^{232}U
^{232,236}U	14	4.4	2357	5.5×10^{-10}	4.9×10^{-3}	0.64	0.344	4.95	0.36	2.5×10^{-9}	4.3×10^{-2}	3.78
^{232,238}U	29	3.0	2821	1.7×10^{-9}	3.5×10^{-3}	0.52	0.339	3.94	0.16	9.0×10^{-9}	4.0×10^{-2}	5.10
^{234,235}U	26	3.2	2576	2.0×10^{-9}	5.2×10^{-3}	0.66	0.362	4.50	0.28	8.8×10^{-9}	4.4×10^{-2}	3.80
^{235,236}U	32	1.4	3094	1.8×10^{-9}	2.9×10^{-3}	0.49	0.368	3.41	0.06	9.9×10^{-9}	3.5×10^{-2}	6.29

These examples show that the best matched abundance ratio cascade is a cascade with key components ^{232,236}U. Moreover, the product flow of the ordinary cascade contains the minimal quantity of the impurity component ^{236}U. The total flow ΣL in this M*-cascade is significantly lower as compared to other cases.

Table 5 shows the isotopic compositions of primary product flows of the matched abundance ratio cascades, which are the best in terms of the flow *P*2. The parameters specified meet the ASTM C 996–10 requirements set for the low enriched uranium.

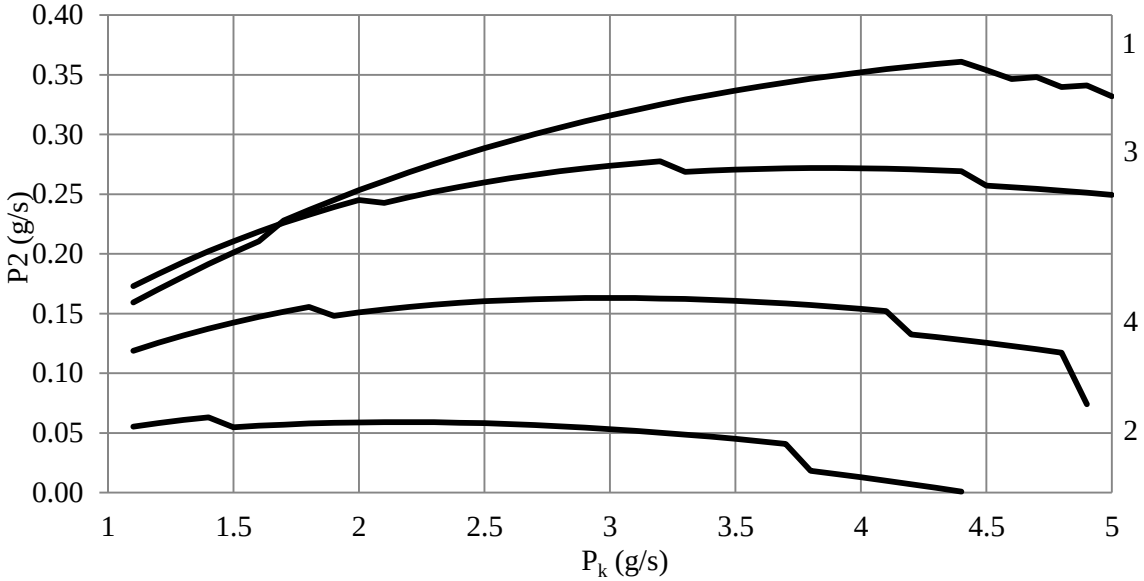

Figure 3. Product flow of ordinary cascade in function of additional product flow of M*-cascade: 1 – key components ^{232,236}U; 2 – key components ^{232,238}U; 3 – key components ^{234,235}U; 4 – key components ^{235,236}U.

Table 5. Parameters of the primary product flows of M*-cascades with maximal product flows *P*2.

MARC	C_i^P(%)				*P*(g/s)	*P*2 (g/s)
	^{232}U	^{234}U	^{235}U	^{236}U		
^{232,236}U	1.65×10^{-6}	0.91	0.36	1.128	1.65×10^{-6}	0.146
^{232,238}U	9.04×10^{-7}	1.65	0.16	1.068	9.04×10^{-7}	0.090
^{234,235}U	1.22×10^{-6}	1.23	0.28	1.109	1.22×10^{-6}	0.115
^{235,236}U	6.73×10^{-7}	2.22	0.06	1.034	6.73×10^{-7}	0.070

4. Conclusion

The method developed allows purifying the regenerated uranium to remove the impurity isotopes $^{232, 234}$U from the additional product flow of the matched abundance ratio cascade with a concentration of the ^{235}U in the product flow up to 5 %. The purified product can be then enriched in another cascade while meeting the requirements of ASTM C 996–10 set for these isotopes in the low enriched commercial uranium. As for achieving the maximum flow of the enriched and purified uranium, the best cascade is the M*-cascade with key components ^{232,236}U.

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